Argonne National Laboratory

INTERNATIONAL COMPARISON OF

^{56}Mn ACTIVITY IN 1968

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INTERNATIONAL COMPARISON OF
^{56}Mn ACTIVITY IN 1968

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ABSTRACT

The absolute radioassay of liquid samples of ^{56}Mn was carried out by six laboratories in Europe and North America in September 1968. Aside from frequent application in a variety of nuclear-reactor irradiations, ^{56}Mn is the basic activity produced in the manganese bath system of neutron-source calibration, which in turn is directly connected with some determinations of the neutron yield from fission. Because of possible related discrepancies in nuclear data, a limited intercomparison was organized by the National Physical Laboratory of England. Although the 2.6-h half-life places a premium on rapid transportation and processing, six laboratories were able to participate simultaneously. All participants calibrated aliquots of the samples through a coincidence technique. The range of values obtained was 0.6%, and the rms error was 0.1%. Despite the narrow range of results, a bias due to calibration technique is possible. The 4π liquid-scintillation/gamma-coincidence method appears to yield disintegration values about 0.3% higher than the 4π proportional-counter gamma-coincidence technique; however, use of a half-life 0.2% lower than the reference value of 2.578 h eliminates part of this apparent bias in counting method. The coincidence data computer program COINC was also tested and found to produce results consistent with data-processing formulas applied at the other laboratories.

I. INTRODUCTION

The absolute detection of ^{56}Mn activity is a cornerstone of the manganese-bath process for determination of neutron-source strengths. Absolute neutron-source rates, in turn, provide either essential or supporting data for the neutron yield in fission (ν) determined by current methods. Because substantial differences have existed in various published measurements of $\nu(^{252}$Cf$)$, a search for possible systematic errors has indicated the need for verification of the assumed ability of various laboratories to calibrate independently with ^{56}Mn.

Accordingly, a limited intercomparison was organized by the National Physical Laboratory, with samples distributed to interested

laboratories. Although the favorable energy characteristics of ^{56}Mn make this isotope excellent for coincidence counting, its short half-life (approximately 2.6 h) caused, as expected, difficulties in distribution.

The numerical results of the comparison are compiled in Table I, which is a summary of findings. Additional supporting information has been included in appendixes contributed by each participating laboratory.

II. SUMMARY

Despite the vicissitudes of transportation and rapid handling required to prepare samples of ^{56}Mn activity for coincidence counting, information of considerable interest has been developed in this comparison. Of six laboratories that received samples, all calibrated by the 4π β/γ coincidence method; four laboratories carried out beta detection with proportional counters, while three of the six used liquid scintillators; one of these latter three applied a liquid-scintillation method without coincidence.

The unweighted average of all results has an rms spread of 0.1%; the range of values is 0.6%. Table I summarizes the data.

TABLE I. ^{56}Mn Intercomparison
Reference Time: 2000 BST[a] on September 11, 1968
$\lambda = 7.469 \times 10^{-5}$ sec^{-1} (= 2.578 h)

	ANL	AECL	BIPM	IKO	BCMN	NPL	Average	Range
Activity Comparison								
Activity reported by Laboratory, dps/mg	2534.7	2529.2	2526.7	2537.8	2526.1[b]	2523.0	2529.6	0.6%
Total error, dps/mg	±5.0	±2.8	±8.0	±6.0	±10.0	±10.0	±2.5	
Standard error of mean, %	0.1	0.04	0.03		0.07	0.02		
Rms error, %	0.33			0.23				
Systematic error, %	0.1	0.06	0.3		0.2	0.4	0.09	
Confidence interval for total error	1σ				3σ$_r$ + σ$_s$	2σ		
Corrections								
Buoyancy, %	0.1	0.1	0.1	None	0.1	0.1	0.1	
Contaminant at reference time, %	0.1-0.2	<0.001	Possible		<0.005	<0.01		
Mean time of measurement after reference time, h	7	12	14	6	7	0		
Average Efficiencies								
Gamma, %	1.7	6.3	3.5	2.8	30/15	13		
Beta, %	98	97.5	98.5	95	99.0/98.5	99.2		
Method Comparison								
4πβ proportional counter/gamma coincidence		2529.2	2526.4		2524.7	2523.0	2525.9	
Standard error, sec^{-1}mg^{-1}		±1.0	±0.7		±1.4	±0.5	±1.5	} 0.35%
4πβ liquid scintillator/gamma coincidence	2534.7			2537.8	2531.7[c]		2534.7	
Standard error, sec^{-1}mg^{-1}	±2.5			±6.0	±2.5		±1.5	
Calculation Comparison								
Calculation by ANL COINC program		d	2523.6	2530.5	2526.2	d		} 0.02-0.12%
Calculation by measuring Laboratory		d	2526.7	2531.8	2526.6	d		
Half-life = 2.573 h[e]	2544.0	2545.2	2545.4	2545.3	2535.3	2523.0	2539.8	0.9%

[a] British standard time.
[b] Adjusted to $\lambda = 7.469 \times 10^{-5}$ sec^{-1}.
[c] Not operated in coincidence mode.
[d] Agreement within 0.02% between COINC and measuring Laboratory.
[e] Reported activity (top line) adjusted according to mean time of measurement after reference time.

One discrepancy is apparent. In general, the liquid-scintillator results are 0.35% higher than the proportional-counter values.

Supplementary investigations so far have failed to develop a supportable explanation for any bias between the proportional-counter and liquid-scintillator outcomes. Using the lowest of the published accurate measurements for the half-life of ^{56}Mn (2.573 h) in place of the reference value (2.578 h) partially eliminates the discrepancy.

Although results consistent with the remaining participants in this comparison were achieved by ANL (which used the ampoule method of delivery for the first time), it was discovered that a significant systematic discrepancy was connected with an earlier ANL practice of measuring aliquots by weighing vials into which the solution was delivered.

The various calculational procedures applied at each laboratory create computational differences of 0.12% at most.

Table II contains a tabulation of nuclear characteristics of ^{56}Mn, and Fig. 1 is the decay scheme.

TABLE II. Nuclear Characteristics of ^{56}Mn

Half-life, h	Confidence Limits	Reported by
2.573 ± 0.004	σ	Reynolds, Emery and Wyatt[1]
2.578 ± 0.008	Estimate of overall error	Goodier[2]
2.578 ± 0.009	3σ + sum of estimated systematic	Lagoutine, LeGallic, and Legrand[3]
2.591 ± 0.008	σ	Liskien and Paulsen[4]
2.576 ± 0.002	σ	Bartholomew et al.[5]
2.5764 ± 0.0012	3σ	Taylor and Merritt[6]
2.5782 ± 0.0005	σ + estimated systematic	Huynh[7]
2.574 ± 0.003	σ + estimated systematic	Lockett and Thomas[8]

γ/dis		
1.422		Chasman and Ristinen[9]
1.529		Bienlein and Dinter[10]
1.42 ± 0.01		Spernol (this report)

Main Beta Groups		Bienlein and Dinter[10]
End-point Energy, keV	Intensity, %	
2838	47	
1028	34	
718	18	
300	1	

Primary Gamma Rays		Chasman and Ristinen[9]
846.64	100	
1811.2	27	
2112.4	14	
2523.6	1	
Sum of 5 others	1	

Angular Correlation		
$W(\theta) = 1 + 0.022 \cos^2 \theta$ (2838-keV beta only)		Lobashov and Nazerenko[11]

Internal Conversion		
<0.01%		Rose[12]

Fig. 1

Decay Scheme for ^{56}Mn (adapted from Chasman and Ristinen[9] and Bienlein and Dinter[10]). ANL Neg. No. 113-2544 Rev. 1.

Participants in the comparison are listed in Table III.

TABLE III. Participating Laboratories

Designation	Laboratory	Participants
AECL	Chalk River Nuclear Laboratories Atomic Energy of Canada Limited Chalk River, Ontario, Canada	J. G. V. Taylor and J. S. Merritt General Physics Branch
ANL	Argonne National Laboratory Argonne, Illinois 60439, U.S.A.	A. DeVolpi, R. J. Armani, and K. G. Porges. Reactor Physics Division
BCMN	Bureau Central de Mesures Nucleaires EURATOM, Geel, Belgium	E. DeRoost, E. Funck, A. Spernol, W. van der Eijk, and R. Vaninbroukx
BIPM	Bureau International des Poids et Mesures Pavillon de Breteuil F-92 Sèvres, France	A. Rytz, V. Naggiar, J. W. Müller, and V. D. Huynh
IKO	Instituut voor Kernphysisch Onderzoek Ooster Ringdijk 18, Amsterdam, The Netherlands	B. J. Mijnheer and G. A. Brinkman
NPL	National Physical Laboratory Teddington, Middlesex, England	A. Williams and I. W. Goodier Division of Radiation Science

III. IRRADIATION AND DISTRIBUTION OF SAMPLE

The National Physical Laboratory undertook to irradiate and distribute vials containing about 1 ml of solution. Monoisotopic manganese metal was irradiated and then dissolved to produce $MnCl_2$ in 0.1 N HCl at a carrier concentration of 2 mg/g and a radioactive concentration of approximately 70 μCi/g at the reference time (2000 BST, September 11, 1968). One or two numbered vials were sent to each of nine laboratories; two of these laboratories did not conduct an absolute calibration, and one did not receive the sample in time to make the measurements.

Except for the sample to the National Research Council (Canada) which went astray, all were received with adequate time for dilution and preparation, although there were some delays, as indicated in Table I.

IV. DISCUSSION

A. Primary Results

Table I has all the data compiled. Although an average of all results, each given equal weight, results in an rms deviation of only 0.1%, there are some noticeable systematic effects. In view of the difficulties arising from the short half-life and the widespread transportation uncertainties, the degree of agreement may be comparable with that expected from an isotope with energetic beta and gamma emission and an essentially ideal decay scheme.

The most prominent effect to appear is a relatively consistent discrepancy involving the method of calibration. This will be discussed further in Section C below.

The standard errors of the mean for each laboratory are less than 0.1%. Systematic errors have been chosen on a nonuniform basis, so that the total error is dominated generally by the systematic error. In some instances, the total error reflects confidence intervals greater than one standard deviation. Since it is difficult to assess systematic errors on unknown possibilities, comparison of quoted systematic errors is not too meaningful. Some laboratories have included errors that affect all data uniformly (such as half-life), but do not affect an interpretation of the intercomparison.

The magnitude of the contaminant remains a matter of contention. ANL measured the long-lived contaminant under actual operating conditions for all three data channels. Some of the other laboratories measured only the gamma-ray or beta effect; others did not follow the activity for as long a period. That there was a contaminant and that its half-life was in the order of 15 h seems agreed; but the relative effect in each detection channel, referred to specific activity at issue time, is considered to range from 0.001 to 0.2%. Although the effect of the contaminant will depend on the cutoff energy of the beta detector, on the upper and lower windows of the gamma channel, and, perhaps, on the beta-detection medium one does not expect 0.2% spread in the extrapolated contaminant.

B. Sample Preparation

All participants delivered aqueous samples through plastic ampoules, which were weighed to determine the aliquot by weight difference. Even the delivery of alcohol-based solutions appears to be accurate by this "pycnometer" method. As a consequence of a search for some inexplicable small discrepancies, a serious discrepancy in a previously used ANL procedure of postdelivery weighing of vials was discovered.

Corrections for buoyancy of about 0.1% were generally applied. In the catalog of noticeable differences between the liquid-scintillation and proportional-counter techniques, it is observed that deliveries for the liquid scintillators are into essentially closed vials, while deliveries for proportional-counter foils are done in open environments. However, the prospect for activity losses in this situation seems rather remote.

There was no evidence that reducing agents were needed to maintain the stability of the activity already incorporated in an acidic carrier.

C. Counting Techniques

Of the six participating laboratories, four used the proportional-counter coincidence method. The other two used the liquid-scintillation coincidence method exclusively, while BCMN used the proportional counter in coincidence and an absolute liquid-scintillator counter without coincidence. Three variations of the scintillator method were applied by IKO,* with all data combined in a weighted average.

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* Instituut voor Kernphysisch Onderzoek.

A wide variety of operational conditions are represented in this ^{56}Mn comparison, in terms of deadtimes, coincidence-resolving times, and mechanical arrangement. With regard to the difference between the two primary methods, there appears to be no connection with these specific operational circumstances. On the other hand, there is perhaps some correlation with gamma-ray efficiency and with beta sensitivity. The existence of this discrepancy is credible if the errors associated with each result are ignored, although there is support from BCMN* observations of a similar trend in the past. Table I indicates that the proportional-counter results cluster uniformly around an average that is markedly less than the liquid-scintillation data. On the other hand, the range of proportional-counter data (0.25%) is the same as the internal range of the liquid-scintillator data, and not much different from the gap between the two methods (0.3%).

At first glance, one supposition would be that the liquid-scintillator method may be overcounting due to afterpulses. This does not appear to be the case, as deadtimes applied are generally sufficient to suppress satellites generated from ion feedback in photomultiplier tubes. In addition, ANL rechecked its equipment with paralysis times going up to 75 μsec. Since liquid-scintillator systems have relatively high beta-detection thresholds (compared to proportional counters), there is a further inherent degree of afterpulse rejection. The two-tube coincidence requirement for liquid-scintillation detection of betas (as at ANL) also tends to exclude phosphorescence as a source of spurious counts in the beta channel.

The other prime suspect is the gamma sensitivity of the liquid scintillator; empirical data using ^{56}Mn seem to exclude such a systematic deficit.

In the search for possible sources of discrepancy, effects were sought that were unique to ^{56}Mn and that tended to exaggerate the physical differences between the liquid scintillators and the proportional counters. One such effect is sensitivity to beta-gamma correlation: By virtue of its design, the proportional counter can be expected to be sensitive to the observed correlation in ^{56}Mn. The upper limit to this effect was calculated to be 0.1%, although an angular-correlation experiment would be more convincing.

Another possibly exaggerated difference is connected with the much higher sensitivity of the proportional counter to low-energy electrons and X rays. Extrapolation curves obtained with ^{56}Mn as the radioactive agent help resolve the question of possible gamma-gamma coincidence bias. Results from AECL and NPL (Appendixes A and F) show that this is not responsible for the overriding discrepancy.

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*Central Bureau for Nuclear Measurements.

At this writing, there is no supportable reason for discounting either method of calibration; further investigation is needed. Laboratories that have both proportional-counter and liquid-scintillator coincidence systems are best suited for comparative work with ^{56}Mn.

D. Half-life

An appropriate half-life of 2.578 h was chosen as a common factor in the calibration. Data in Table II suggest that a value around 2.577 or 2.578 h is reasonable. The choice of either of these two half-lives causes a difference of only about 0.1%. The largest difference being associated with a 12-h mean counting time delay.

However, applying the half-life of 2.573 h reported by Reynolds, Emery, and Wyatt[1] to the mean time of measurement after reference time (from Table I) appears to eliminate the technique-dependent bias partially. The six activities become as shown in the bottom line of Table I.

An important factor of this calibration with ^{56}Mn is the constraint caused by its relatively rapid decay. Resolution of the discrepancy among the carefully measured half-lives listed in Table II is important in terms of understanding the origin of the possible bias presented in this ^{56}Mn calibration. The effect of the half-life uncertainty upon neutron-source calibration, though, is likely to be negligible, since typical manganese-bath standardization procedure calls for nearly simultaneous counting of activity dissolved in the bath against beta-gamma detection of gravimetrically related samples.

E. Computational Method

One objective of this comparison of ^{56}Mn activity was to take ad - vantage of this first opportunity to exchange calculations on common data. The published program COINC was used as a basis, with the results shown in Table I. Since not all the data were duplicated for treatment by COINC, the numbers given will not agree with the final laboratory result. However, the differences given vertically are a measure of the effect caused by the calculational method upon common data. No more than 0.12% difference develops between COINC and the method normally applied by the measuring laboratory. Count rates in the beta channel ranged from less than 1000 to 12,000 counts/sec.

Since COINC is based on derivations by Gandy,[13] and most of the other laboratories have applied the earlier Campion model,[14] it appears that there is little difference in the methods when applied to ^{56}Mn. As a result, we can consider the fully published code COINC to be verified for a wide range of applications.

APPENDIX A

Atomic Energy of Canada Limited (AECL)

1. Sample Preparation

The samples did not reach Chalk River until almost 10 hours after the reference time. Six sources of 42-88 mg of the undiluted solution were weighed from a polyethylene pycnometer (AECL-2679) onto standard gold-coated VYNS source mounts, dried under a heat lamp, and sandwiched with a similar film to give a total source-mount thickness of ~40 μg cm^{-2}. The buoyancy correction, which was essentially the same for all sources, was applied to the final mean value of the disintegration rate rather than to the individual weights.

2. Counting

The samples and three blank source mounts for background measurements were counted in a 4π $\beta(pc)$-γ coincidence system with automatic sample changer and readout. The stainless steel pillbox type 4π beta flow counter was 3.8 x 2.0 cm high (each half) with 0.013-mm stainless steel anodes operated at 2600 V using CH$_4$ gas. The gamma rays were detected in two 7.6 x 7.6-cm NaI(Tl) crystals. Single-channel analyzers accepted a window around the 847-keV photopeak from each detector, and their outputs were connected in parallel at the input of the deadtime shaper. (Actually, two independent sets of electronics were used in parallel and the data recorded simultaneously. The second system was set with wide windows in the gamma channel, which gave a higher counting rate but a much poorer signal-to-background ratio. The results were consistent with those from the first system, but were not used to get the final result because they were less precise, due to background fluctuations, and less accurate, due to the relatively large uncertainty in the much larger efficiency-dependent correction associated with the wider window.)

The deadtimes in the beta and gamma channels were 2.00 and 2.03 μsec, and the total coincidence resolving time, $\tau_r(\beta) + \tau_r(\gamma)$, was 1.392 μsec. The average channel delays were set equal. All counts were for a preset interval of 1000 sec. A frequency standard provided the time base for the timer and the digital clock. The channel counts and the time for each measurement were typed out and punched on paper tape.

All the samples were followed for 36 h; the strongest one was then followed for a further 48 h and was remeasured during the next week. Only the first 5 h of counting data were used to determine the activity; the later results were used to estimate the long-lived contaminants. When the counting started, the strongest source was about 9000 dps. Background corrections averaged ~0.01, 0.3, and 0.003% in the beta, gamma, and coincidence channels.

3. Calculations

Disintegration rates were obtained from the paper-tape output using a computer program BEGAMCOP based on the more exact of the formulas derived by Campion.[14] The correction for decay during the counting interval is not included in the program, but was applied to the final mean-disintegration-rate value since it was the same for all measurements. Analysis at AECL indicates that this program gives results about 0.01% lower than the ANL COINC program for the average source-disintegration rates of about 5×10^3.

4. Results

The average correction made for observed long-lived contaminants was 0.05%. The principal contaminant appeared to have a half-life of about 15 h and was assumed to be ^{24}Na, in which case it would have been about 0.002% of the ^{56}Mn activity at the reference time. Sodium-24 was felt to be the most likely contaminant, because, even with the window around the 847-keV photopeak only, it showed up clearly in the coincidence as well as the beta channel (it was masked by background fluctuations in the gamma channel), and because ^{24}Na is a commonly observed contaminant in reactor-produced activities. If the contaminant were ^{64}Cu, as suggested by the ANL data, it could have been as much as 0.005% at the reference time without seriously conflicting with the AECL data. No conceivable hypothesis could bring the level of the ~15-h contaminant in the AECL sample within an order of magnitude of the 0.1-0.2% observed in the ANL sample.

The observed contaminant with a half-life $\gg$15 h was more than an order of magnitude less than the 15-h activity at the start of counting and was not identified. After the observed long-lived components were sub-tracted, the remaining activity appeared to decay with a half-life slightly longer than 2.578 h; the deviations were less than the statistical errors, but seemed to show a discernible trend. (A similar trend was observed by the BIPM; their data were best fitted by a half-life of 2.583 h.) Later, the half-life for a freshly prepared ^{56}Mn sample showing no observable impuri-ties was computed from high-pressure 4π gamma ion-chamber data to be $[2.5764 \pm 0.0012 \ (3\sigma)]$ h in agreement with an earlier Chalk River value of 2.576 ± 0.002 h. In addition, the data of Table I indicate a possible correla-tion between higher answers and later counting times for the $4\pi \ \beta(\text{pc})$-γ results. Together these observations might indicate the presence of another weak contaminant with a half-life of a few hours. On the other hand, the NPL measurements, which included the time interval immediately following the reference time, showed no such effect.

Table I lists the final AECL value for the ^{56}Mn activity, together with the statistical and estimated systematic errors.

APPENDIX B

Argonne National Laboratory (ANL)

1. Sample Preparation

The samples were received on schedule at ANL. After activity monitoring, the solution from one vial was drawn into a preweighed polethylene ampoule. This solution was then delivered into a preweighed 50-ml flask, subsequently filled with water. A few drops of peroxide were also added to the solution. From the diluted solution, a second polyethylene ampoule was filled; approximately 50 μl of this solution were transferred to each of five liquid-scintillation vials. The vials contained about 200 μl of ethanol wetting the glass before the active solution was added.

The remainder of the solution in the 50-ml flask was poured into the manganese bath for a parallel equipment calibration. All aliquot weights for the liquid-scintillation vials were determined by weight differences of the ampoule; the dilution factor was derived from the weight of liquid transferred from the stock solution via an ampoule; and the amount of diluted solution inserted in the manganese bath was gravimetrically determined. For buoyancy effects, a correction of about 0.1% was applied.

Each liquid-scintillation vial was filled with a dioxane-based scintillator and then capped.

2. Counting Procedure

The five samples were counted in turn for 5 min, and the cycle was repeated about four times over an interval of almost 3 h, starting approximately 5.5 h after reference time. The next morning, one of the samples was recounted by automatic apparatus for about seven more hours.

The dilution factor of 20 provided activities of 1100-1400 dps, which is a factor of two or three less than normally employed here in routine manganese-bath calibrations. The overdilution resulted from an error in our activity assay, which concurred with the provisional notification accompanying the sample, although actual specific activity was less than half the provisional estimate.

Because of the relatively low magnitude of sample activity, count rates were more sensitive to background influences than usual. Initial background components were: beta, 0.1%; gamma, 4%; and coincidence, 0.2%.

Detection of betas is based on two low-noise photomultiphiers coupled to the faces of the liquid-scintillation vial, operating in coincidence at a level that produces a tritium efficiency of about 10%.

Gamma-ray detection is obtained with a 7.6-cm sodium iodide crystal. A rather broad window surrounding the 847-keV line runs from above 500 keV to below 1500 keV.

A deadtime of 14 μsec is applied in the beta channel and 5.2 μsec in the gamma side. The total coincidence resolving time is 0.375 μsec.

Data from all channels--as well as the cumulative and count times-- were recorded by an oscillator-driven automatic-card-punch apparatus.

3. Results

The ANL average specific activity reported in Table I is a weighted mean of the five samples, each counted with a standard deviation of 0.2-0.3%. Weights were assigned according to the square of the standard deviations, and the standard deviation of the mean was computed in the usual manner from the inverse sum of weights. Table IV is a summary of ANL data.

TABLE IV. ANL Summary
2.578 h--2000 BST

Sample No. :	1	2	3	4	5	
Weighted mean, dps/mg	2564.9	2530.1	2542.8	2517.4	2545.0	
Standard deviation, dps/mg	6.6	5.5	7.1	7.2	5.9	
ϵ_β, %	97.3	98.8	98.6	98.7	97.0	
$\dfrac{1 - \epsilon_\beta}{\epsilon_\beta}$	0.028	0.012	0.014	0.013	0.031	
Extrapolation factors	0.9970	0.9987	0.9985	0.9986	0.9967	
Disintegration rate, dps/mg	2557.2	2526.8	2539.0	2513.9	2536.6	
Weight, $(mg/dps)^2$	0.023	0.033	0.020	0.019	0.029	
Weighted mean, dps/mg						2534.7
Standard deviation, dps/mg						2.8
Standard-deviation, percent error						(0.11)
Rms error, dps/mg						8.4
Rms percent error						(0.33)
Systematic error, %						0.1
Total error, % (sum of standard deviation and systematic error)					0.2 (CI = 0.68)	

Efficiencies for beta detection (ϵ_β) were 97-98%, which is our normal range. Gamma-channel efficiency is almost 1.7%. A correction of about 0.2% was applied to each sample to account for the interaction between the decay scheme and the finite gamma sensitivity of the beta channel in accord with a linear extrapolation as a function of $(1 - \epsilon_\beta)/\epsilon_\beta$.

A relatively substantial spread in our result (several tenths of a percent) arises from choice of sample background. In routine calibrations, we sometimes find that the sample background exceeds the blank liquid-scintillator background; this was also found to be true for this issue. The background for the sample was deduced from data obtained in the interval from 70,000 to 95,000 sec after reference time. To rectify the data in this period, the derived beta background was 30% above normal; gamma background, 5% above normal; and coincidence, three times normal. The largest uncertainty, though, is introduced by extrapolation of these backgrounds to the initial counting period. A decay period of 15 h was noted, which is consistent with either ^{24}Na or ^{64}Cu contamination. The extrapolation of this excess long-lived activity, which appears in all channels, results in our numbers being several tenths of a percent less than unextrapolated data.

The estimated systematic error does not take into account any inherent deficiencies of the liquid-scintillation method. Errors for the background subtraction were sufficiently liberal (40% in the beta channel, 6% in gamma, and 40% in coincidence) to account for uncertainty in the extrapolation; these background errors are already reflected in the standard deviation for each sample. The program COINC, which was used for computation of results, also compounds errors in half-life, deadtimes, resolving times, and count times.

4. Evaluation and Supplementary Investigations

When it appeared that a systematic difference (of about 0.3%) might exist between two methods of coincidence counting of ^{56}Mn, an investigation was launched into some of the more prominent hypotheses. We have looked for special complexities introduced by the decay scheme, but found no probable cause for the difference.

Particular attention was paid to the possibility of residual satellite pulse effects in the liquid-scintillation method. Our results obtained from imposing deadtimes up to 75 μsec (which far exceed ion feedback delays in phototubes), indicate no detectable dependence on afterpulses, as shown in Table V.

Several factors tend to exclude phosphorescence as a possible cause of excess counts in the ANL equipment: the two-tube coincidence requirement, the relatively high beta channel bias, the absence of observable effects in this and prior ^{56}Mn radioassays followed for lengthy periods, and the failure of phosphorescence to appear in tests with weak beta emitters with much longer half-lives.

Since the same type of delivery method was used by all participants in this calibration, the deficiency probably is not related to delivery technique (even though there is a procedural difference regarding expulsion to an open planchet compared to a closed liquid-scintillator vial).

TABLE V. ANL ^{56}Mn Sample 204

(January 16, 1969)

Deadtime, μsec	No. of 10-min Counts	Disintegration Rate, dps/mg
14	2	73.37 ± 0.22
25	2	73.25 ± 0.34
50	2	73.08 ± 0.51
75	19	73.31 ± 0.15
	19	73.08 ± 0.24
	19	73.31 ± 0.57
	20	74.75 ± 1.33
	16	73.23 ± 3.42

One unique contrast between liquid-scintillator and proportional-counter methods is associated with angular anisotropy. The liquid scintillator may be considered, essentially, a true 4π beta detector, even allowing for wall effects. On the other hand, the proportional-counter method has a potentially oriented response, which diminishes along the foil plane, with the gamma detector ordinarily aligned along the high-efficiency normal axis. Despite rather extreme assumptions regarding this response, from calculations based on the known anisotropy of emission for the allowed 2.86-MeV beta (in Table II), an upper limit of 0.1% is computed. However, actual measurements with the sodium iodide crystals arranged parallel and perpendicular to the foil plane, under a variety of sample conditions, would be required to fully satisfy this speculation for ^{56}Mn.

Another prominent difference between the two techniques emanates from gamma sensitivity of the beta detector. Generally, liquid-scintillator response to gamma rays is an order of magnitude larger than for proportional counters. For ^{56}Mn, a sensitivity of 9.6% was used by IKO, and this is consistent with our evaluation in ANL geometry. However, since liquid-scintillator efficiencies run up to 98%, the net effect upon the disintegration rate is expected to be only 0.2% which is in agreement with linear extrapolations from experimental data. For the energetic gamma rays of ^{56}Mn, estimates for gamma sensitivity applied to the proportional counters are about 0.5%.[15] This correction alone opposes matching of the discrepancy, but there is another complexity that would increase the proportional counter result with little corresponding effect on liquid-scintillation data. This is the gamma-gamma contribution to the coincidence channel, which, as emphasized by BCMN,[16] is highly sensitive to the effective discrimination level. Since proportional counter systems typically have much lower cutoff energies in beta detection than the liquid scintillators, the possibility of a gamma-gamma coincidence during beta-event nondetection cannot be ignored. Gamma efficiencies and solid angles are rather large in some cases of this ^{56}Mn intercalibration. In any event, calculations based on empirical data do not lend credence to an effect larger than 0.1%.

As it stands, we have derived no supportable explanation that would suggest that the liquid-scintillator data, which are backed by extensive empirical evidence, are incorrectly high, nor have we found sufficient effects to cause the proportional-counter data to be incorrectly low.

5. Appearance of a Gravimetric-sampling Deficiency

One objective of this comparison was to assist in the isolation of discrepancies that apparently have existed in neutron-source calibration. To some extent this has been accomplished, because a serious gravimetric-sampling discrepancy at ANL was subsequently noted affecting a 1.5-yr period of our data.

Up through 1966, comparative calibration of our manganese bath was effected by volumetric deliveries from an ultramicropipette. Afterward, we changed to a gravimetric technique of weighing delivered aliquots in promptly capped vials. Extrapolation data indicated that leakage and evaporation were negligible. We now find that upon delivery of ethanol solutions to the vial, a rapid initial evaporation occurs upon contact with the wall of the vial at room temperature, resulting in loss of about 1 mg of solution.

This explains several inconsistencies that have appeared in our previous data. For previous intercomparisons with NRC and NBS in 1965, we used gravimetric delivery, resulting in a 2.8% average discrepancy. Two percent of this can now be accounted for by the weight loss after delivery; the remainder is associated with the existing systematic difference between liquid-scintillator and proportional-counter techniques, with our neglect of the 0.2% extrapolation effect at the time, and with possibly some afterpulse sensitivity for the coincidence-system parameters then used.

For the present comparison, we shifted to the ampoule weight-difference method following J. G. V. Taylor's suggestion. We also supplemented our data with what appeared to be a replication of our normal delivery technique, namely weighing of vials after delivery. Only a 0.2% loss was apparent, but the true loss that occurs under normal conditions was masked by our introduction of alcohol to the bottom of the vial before addition of the water-based activity. As a result, the vial was preweighed (with some alcohol) after the prompt contact loss of alcohol. When the aqueous activity was then added to the wetted vial, only the small 0.2% loss followed.

The net result of this revelation is to cause some of our neutron-source rates to be lowered (depending on their weighting with respect to the overall period of calibration), removing one fixed discrepancy that had existed between NPL and ANL data.

We were stimulated to follow through in this investigation initially in terms of the 0.2% discrepancy that appeared in the intercalibration and subsequently verified by NPL. Application of a correction for this loss also accounts for a previously unresolved arbitrary normalization of neutron-source data in 1967. In retrospect, we can also now understand why certain local calibrations with proportional-counter equipment failed to uncover this deficiency: It was masked by variations in the delivery procedure corresponding to the type of intercalibration.

APPENDIX C

Central Bureau for Nuclear Measurements (BCMN)

1. Sample Preparation

From the original solution A (ampoule No. 16), three sources were prepared onto thin gold-coated VYNS foils. The drop weights were determined with the pycnometer method using a plastic pycnometer.[17] The rest of solution A, about 0.9 ml, was diluted quantitatively with about the same amount of 0.1N HCl and divided into two equal parts, B_1 and B_2. B_1 was diluted immediately in the same way, and from the final solution C_1, two pycnometers were filled. From each pycnometer, three sources were prepared onto thin gold-coated VYNS foils. Thereafter, two pycnometers were filled from solution B_2, and again three sources prepared onto gold-coated VYNS foils from both of them. To all foil sources, one drop of an aqueous 0.01% Ludox SM solution was added. The drops were dried in a nitrogen stream at about 35-40°C.

The remainder of solution B_2 was diluted 1:5 with 1N HCl. From the final solution C_2, three sources for liquid-scintillation counting were prepared.

A Sartorius MPR-5 microbalance was used for the drop-weight determinations with the pycnometer method. The weights of the liquid-scintillator sources were determined both by the pycnometer method and by an extrapolation method. In the last case, a Mettler B6 semimicrobalance was used.

2. Counting Procedure

All foil sources were measured with the calibrated gamma spectrometer.[18] Most of the sources have been measured several times using the $4\pi\,\beta/\gamma$ coincidence method. Two similar $4\pi\,\beta/\gamma$ counters were used, one employing rather fast electronics with deadtimes of 0.6 and 0.8 μsec and coincidence resolution of 0.2 μsec, and the other with slow electronics with deadtimes of about 5 μsec and coincidence resolution of about 1 μsec, but with two chains in parallel. Most of the sources also were measured with the simple $4\pi\,\beta$ method, again using two different counters.

With the liquid-scintillation method,[19] three sources were measured.

In all measurements, the statistical error in counting was kept below 0.1%. All times were taken with calibrated stopwatches relative to a 10^{-8} precise quartz clock.

3. Results and Discussion

The specific activity at reference time measured by the $4\pi\,\beta/\gamma$
2524.8 dps/mg ± 0.4%, by 4π counting

2528.0 dps/mg ± 0.9%, and by liquid-scintillation counting 2531.7 dps/mg ± 1.0%. The weights used are the reciprocal squared errors. The stated errors are maximum error limits calculated as the sum of all individual systematic errors and three times the statistical error. The experimental standard deviations were 0.23% for the 4π β/γ measurements (12 sources), 0.32% for the 4π measurements (nine sources), and 0.15% for the liquid-scintillation measurements (three sources). A factorial analysis showed that a standard deviation of 0.2% (an unusually high figure) had to be attributed to the dilution and the preparation of the foil sources. The results of the measurements with the gamma spectrometer were not used for the activity determination, because the efficiency calculation depends strongly on the decay scheme used. However, we calculated the efficiency for different decay schemes and found a simple relation between it and the total number of gamma rays per decay. From that and the experimental result for the spectrometer activity, the number of gamma rays per decay has been determined to (142 ± 1%), in agreement with a recently published value.[9] The gamma impurity of the solution was measured with a germanium-lithium detector before the activity measurements started and was found to be less than 0.01%. All decay corrections were calculated using the recommended value for the half-life, including the necessary corrections for decay during the measurement. A significant difference of 0.28% was found between all measurements on solutions (A and C_1) and (B_2 and C_2). Because the latter were exposed to air a longer time, a (0.14 ± 0.14%) correction for evaporation was applied to the weights of the sources of the latter solutions.

We are not satisfied with our results, because the activity was too low for a standardization of highest accuracy. We expected 3 ml, and, even worse, the originally scheduled airplane was missed. Since the solution was received late in the evening, measurements were started at midnight. The original specific activity was then down to 20 μCi/g, and we had to work on small amounts of solution. This explains the unusually high standard deviation on the dilution plus source preparation and the significant difference between the means for the different dilutions. All this caused the rather bad accuracy of 0.4%, which had to be attributed to the final result. Under normal conditions, a two-times better figure could be reached.

APPENDIX D

Bureau International des Poids et Mesures (BIPM)

1. Source Preparation

Since the ampoule ordered from NPL arrived several hours later than expected, the time schedule had to be modified and the counting started considerably later than planned originally.

Soon after its arrival, the ampoule was opened and its content sucked into a polythene pycnometer. No dilution was made. About one hour later, the sources were prepared by dispensing a drop of solution onto each of the eight standard supports. The latter consisted of a VYNS film, about 8 μg cm^{-2} thick, gold plated on one side by about 15 μg cm^{-2} and subtended by a stainless steel washer, 0.1 mm thick, with 40- and 16-mm outer and inner diameters respectively. The drop masses were chosen in such a way as to get count rates between 3000 and 5000 beta counts per second from each source. The total source mass was 648 mg. To each source drop, two drops of Ludox SM (10^{-4}) were added for better spreading out. After drying in open air, each source was covered by a similar gold-coated VYNS film, the gold layers forming the outer surfaces of these "sandwiches."

2. Counting Procedure

Each source was counted during two periods of 900 sec in each of the two 4π β(PC)-γ equipments. However, the older set suffered from a considerable gate shift in the gamma channel. Therefore, these results will not be considered here. (For a full description of the counting set, see Ref. 20.) The last and most massive source was counted four times during 900 sec.

3. Calculation of Specific Activity; Corrections Applied

The counting results have been converted into specific activity at the reference time, using two different formulas:

$$SA = \frac{FD\ e^{\lambda(T_0 + T_z)}}{W} \frac{(N_\beta - B_\beta)(N_\gamma - B_\gamma)}{(N_c - B_c - N_{acc})[1 - \tau F(N_c - B_c)]}, \tag{1}$$

with

$$F = \frac{\lambda}{1 - e^{-\lambda T}}$$

and

$$N_c - B_c - N_{acc} = (N_c - B_c) \frac{1 - 2\tau_r\{N_\beta N_\gamma / [T^2 F(N_c - B_c)]\}}{1 - \tau_r(N_\beta + N_\gamma)/T};$$

and

$$SA = \frac{FD\,e^{\lambda(T_0 + T_z)}}{W} \frac{\left[\dfrac{N_\beta}{1 - \tau_\beta'(N_\beta/T)} - B_\beta\right]\left[\dfrac{N_\gamma}{1 - \tau_\gamma'(N_\gamma/T)} - B_\gamma\right]}{\tilde{N}_c}, \qquad (2)$$

with

$$\tilde{N}_c = \frac{N_c\left(1 + \dfrac{\tau_\beta' N_\beta}{T - \tau_\beta' N_\beta} + \dfrac{\tau_\gamma' N_\gamma}{T - \tau_\gamma' N_\gamma}\right) - \dfrac{T\tau_r' N_\beta N_\gamma}{(T - \tau_\beta' N_\beta)(T - \tau_\gamma' N_\gamma)}}{1 - \dfrac{\tau_r'}{2}\left(\dfrac{N_\beta}{T - \tau_\beta' N_\beta} + \dfrac{N_\gamma}{T - \tau_\gamma' N_\gamma}\right) + \tau'\dfrac{N_c}{T}} - B_c.$$

The symbols used in these formulas have the following meanings:

λ decay constant

T_0 0 h of the day following reference time

T_z time at which counting started

T counting time interval

τ deadtime of the beta or gamma channel (whichever is shorter); both deadtimes are supposed to be longer than the deadtime in the coincidence circuit

τ_r coincidence resolution time

W source mass (not corrected for buoyancy and evaporation of solvent)

D dilution factor

N_β observed number of beta counts for time interval T

N_γ observed number of gamma counts for time interval T

N_c observed number of coincidences for time interval T

B_β number of beta-background counts for T

B_γ number of gamma-background counts for T

B_c number of coincidence-background counts for T

$$\left. \begin{array}{l} \tau'_\beta = \tau_\beta(1+a) \\[4pt] \tau'_\gamma = \tau_\gamma(1+a) \\[4pt] \tau' = \tau(1+a) \\[4pt] \tau'_r = \tau_r(1+a) \end{array} \right\} \text{decay-adjusted deadtimes and resolution time}$$

$a = \dfrac{(\lambda T)^2}{12}$ decay adjustment factor (Ref. 21).

Formula 1 is based on a paper by Campion.[14] Formula 2 has been used by DeVolpi et al.[22] and is based on an approximation given by Gandy.[13] Figure 2 shows that, for the present measurements, the results obtained with Eq. 1 are always about 0.1% higher than the ones obtained with Eq. 2.
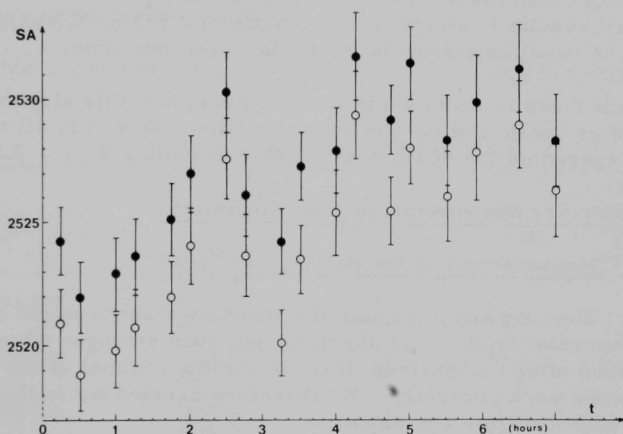

Fig. 2. SA (Specific Activity) in $\sec^{-1}\mathrm{mg}^{-1}$ on September 11, 1968, at 2000 h BST, Calculated Using Formula 1 (full circles) and Formula 2 (open circles) with $T_{1/2} = 2.578$ h. ANL Neg. No. 113-2444.

The computer program included also the determination of the effficiencies, weighted mean, and apparent half-life. The results appear in Section 6 below.

The equations do not take into account all the corrections needed. The following additional corrections have therefore been applied:

a.   Buoyancy had been corrected for by multiplying each source mass by 1.00105. The determination of this factor is explained in Ref. 23.

b.   The evaporation of solvent from the open ampoule and pycnometer gives rise to a correction of 0.01% with the same sign as the buoyancy correction.

c. The decay correction is commented on in Section 4 below.

d. The decay-scheme dependent correction, evaluated according to the procedure proposed by Axton,[21] would be -0.08%. No experimental proof has been obtained, though; thus, this value is treated as a possible systematic error.

4. Appearance of Contamination

Figure 2 illustrates the results of each measurement calculated with both equations. Each pair of subsequent values refers to the same source. The bars correspond to standard errors.

The results, obtained using a half-life of $T_{1/2} = 2.578$ h, show a definite tendency to increase with time. A least-squares fit for the 7-h measuring period results in an apparent half-life of 2.583 h. Clearly this value should not be used for extrapolating to the reference time.

Since there is some evidence for impurities, this slope may be interpreted as due to radioactive contamination. However, all the results have been corrected for decay over 11.25 h assuming $T_{1/2} = 2.578$ h.

5. Supplementary Measurements and Calculations

a. Measurement of the Half-life of ^{56}Mn

The long apparent half-life mentioned above might also be due to an instrumental fault. This suspicion got even stronger when we discovered, soon after comparison, that the gamma channel of the $4\pi \beta/\gamma$ set did not always work correctly. We therefore carried out half-life measurements with the same equipment.

Two samples of ^{56}Mn have been ordered from Saclay at different dates and were measured by different procedures. Table VI gives the details of these experiments from which a value of (2.579 ± 0.001) h (standard error) can be derived. Since this value agrees with the ones found

TABLE VI. Measurements of the Half-life of ^{56}Mn

Sample No.	No. of Sources	Efficiency of the β Counter, %	No. of Measurements per Source	Measuring Time per Source (Total)	Method	Eq.	$T_{1/2}$, h	Standard Error, h
1	1	-	44	900 sec (11 h)	$4\pi \beta$	-	2.5776	0.0006
2	10	98.5	2	900 sec	$4\pi \beta/\gamma$	1	2.5771	0.0008
						2	2.5812	0.0010
2	1	98.4	50	900 sec (12.25 h)	$4\pi \beta$	-	2.5794	0.0006

by other workers, we assume that the fault discovered later had not affected our results of the comparison.

A further measurement carried out in this Laboratory with a different setup suggests a provisional new value for the half-life of (2.5782 ± 0.0005) h (overall error).[7]

b. Recalibration of the Manganese Bath

The remaining 400 mg of the ^{56}Mn solution were introduced into the BIPM $MnSO_4$ bath.[24] The calibration constant obtained with this solution and with the specific activity given in Section 6 was 5325 sec^{-1} per 10^6 dis/sec. In February 1968, this constant was 5323.

6. Summary of the Results, Errors

Table VII summarizes the results obtained from the eight sources, using Eq. 1.

TABLE VII. Summary of BIPM Results

Source No.	Uncorrected Source Mass, mg	Specific Activity (2000 BST, Sept. 11, 1968), $sec^{-1}mg^{-1}$	Standard Error,[25] $sec^{-1}mg^{-1}$	Beta Efficiency, %	T_Z, Start Time of Each 900-sec Measurement
1	45.638	2522.67	1.48	98.7	0800
		2523.36	1.54	98.7	0816
2	40.044	2524.00	1.42	98.8	0715
		2521.71	1.46	98.8	0731
3	57.672	2530.03	1.63	98.6	0930
		2525.88	1.67	98.7	0946
4	56.099	2524.87	1.49	98.7	0845
		2526.76	1.54	98.7	0901
5	81.680	2527.71	1.74	98.2	1100
		2531.54	1.81	98.2	1116
6	108.780	2524.00	1.34	98.4	1015
		2527.07	1.39	98.4	1031
7	111.400	2528.16	1.85	98.1	1231
		2529.71	1.95	98.1	1255
8	146.064	2528.98	1.42	98.4	1145
		2531.30	1.47	98.4	1201
		2531.11	1.81	98.3	1330
		2528.14	1.92	98.3	1400

Uncertainties in the following values or corrections possibly contributed to the overall systematic error and have been estimated according to previous experience:

	Effect on Final Result, %
Deadtime correction	0.05
Background correction	0.03
Accidental coincidences (correction)	0.03
Decay-scheme dependent correction	0.08
Source preparation and weighing	0.10
Timing	0.02
Half-life	0.08
Linear sum	0.39
Adding up in quadrature	0.17

The weighted mean of the specific activity on September 11, 1968, at 2000 BST, is 2526.7 $sec^{-1}mg^{-1}$, with a standard error of 0.7 $sec^{-1}mg^{-1}$, (0.03%) and an estimated overall systematic error of 7.6 $sec^{-1}mg^{-1}$ (0.3%).

APPENDIX E

Instituut voor Kernphysisch Onderzoek (IKO)*

1. Sample Preparation

The undiluted solution was dispensed from a polyethylene ampoule, and the weight of the aliquot was obtained from the weight difference of the pycnometer. For evaluation of the efficiency of the counting arrangement, consisting of three Geiger-Müller counters, about 0.5 g of the solution was poured into a manganese-sulphate bath.[26]

Three types of counting procedures[27] were used in the absolute calibrations:

    a. 4π β/γ Liquid-scintillation Counting

The sample was put into a vial and dissolved in 12-ml p-dioxane containing 4 g/liter PPO, 0.4 g/liter POPOP, and 66 g/liter napthalene.

    b. 4π β/γ Filter-paper-scintillation Counting

The active solution was put onto a disc of filter paper (diameter 2 cm), which was suspended from a thin needle. After being dried with infrared light, the paper was put on top of a plastic disc and wetted with some drops of isopropylbiphenyl containing 4 g/liter PPO and 0.4 g/liter POPOP. The filter paper was covered with a reflector of aluminum foil.

    c. 4π β/γ Plastic-scintillation Counting

The active solution was put onto a 20 x 20 x 0.1-mm plastic scintillator. After evaporation of the water, a second plastic scintillator disc was put on top of the bottom one, sandwiching the activity between the two. No corrections were made for buoyancy effects.

2. Counting Procedure

Beta detection was obtained with a photomultiplier, and gamma detection with a sodium-iodide crystal. The gamma rays were measured integrally above 240 keV. The background during the counting amounted to: beta, 0.01%; gamma, 1%; and coincidence, 0.02%.

The samples were counted at three voltages for the beta-detector multiplier; for each voltage, five discrimination settings were taken. The range of the beta efficiency, ϵ_β, was 0.50-0.90 for the filter papers and 0.75-0.95 for the vials and the plastics. The value of $N = N_\beta N_\gamma / N_c$ was corrected, in the way described by Louwrier,[26] for decay, deadtimes T_β and T_γ and finite resolving times T_β^p and T_γ^p:

$$T_\beta = (1.8 \pm 0.12) \ \mu sec;$$
$$T_\gamma = (3.8 \pm 0.27) \ \mu sec;$$
$$T_\beta^p = (1.0 \pm 0.07) \ \mu sec;$$
$$T_\gamma^p = (0.6 \pm 0.04) \ \mu sec;$$

The corrected values of N were plotted against ϵ_β and extrapolated to $E_\beta = 1.00$ (Fig. 3). After correction for decay and deadtime, the values of N_β were also plotted against the discrimination settings and extrapolated to discrimination setting zero (Fig. 4).
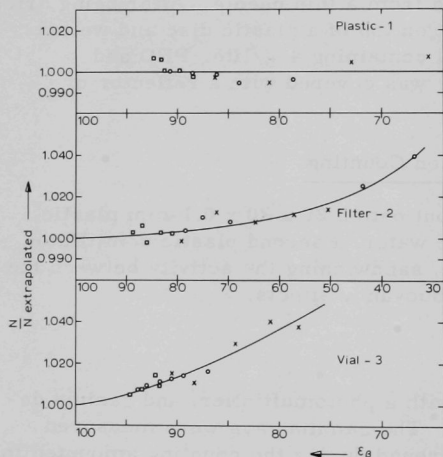

Fig. 3. Dependence of Disintegration Rate upon Beta-channel Efficiency. ANL Neg. No. 113-2445.
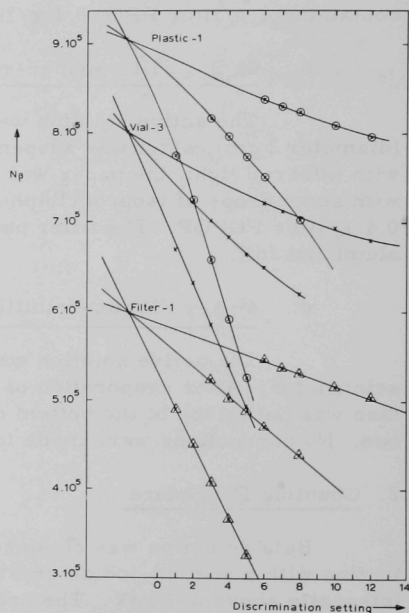

Fig. 4. Dependence of Beta-channel Rate upon Discrimination Setting. ANL Neg. No. 113-2443.

3. Results and Discussion

As is seen from Table VIII, activity values obtained for vial and plastic measurements are higher than those for filter paper. In this connection, 4π beta calibrations in filter-paper discs tend to give low results for beta emitters with a maximum energy less than 1 MeV, and one of the major beta branches in the decay of ^{56}Mn has a maximum energy of only 0.7 MeV. In all 4π beta measurements, extrapolation to $\epsilon_\beta = 1$ introduces some uncertainty, but for our filter-paper measurements this is specially serious, due to the low value of the maximum beta efficiency needed.

TABLE VIII. Summary of IKO ^{56}Mn Calibrations

Method	$4\pi\ \beta$, dps/mg	$4\pi\ \beta/\gamma$, dps/mg
Vial No. 1	2548.3	2538.8
No. 2	2527.8	2534.1
No. 3	2555.7	2533.4
Average	2543.9	2535.4
Filter No. 1	2518.7	2552.4
No. 2	2530.7	2515.1
No. 3	2518.4	2528.6
Average	2522.6	2532.0
Plastic No. 1	2556.7	2545.3
No. 2	2551.4	2555.2
Average	2554.0	2550.2
Overall average	2538.5	2537.8
Standard deviation	6.3	4.6

It seems reasonable to consider that coincidence measurements are more accurate than 4π beta measurements, although the overall averages for the two groups are practically equal.

For our final result, all coincidence measurements were averaged. However, if we would exclude the coincidence measurements with a plastic beta detector, the average would be lowered by less than 0.2%.

APPENDIX F

National Physical Laboratory (NPL)

1. Sample Preparation

The solution from each of four issue-level ampoules was diluted to counting level, approximately 5 μCi/g with 0.1N HCl. The dilution factors were obtained gravimetrically and checked by ionization-chamber measurements.

From each counting-level solution, ten 4π sources, on gold-coated VYNS films, and six ampoules were prepared using glass pycnometers; the source weights were obtained from difference weighings of the pycnometer.

The relative activity of the solution in the issue-level ampoules and the counting-level ampoules was measured, using a sealed high-pressure ionization chamber as an independent check on the dilution factor. Also using the 4π β/γ measurements, a calibration factor for the ionization chamber was obtained. A comparison of this calibration figure with those obtained from previous standardizations gives a consistency check on the measurements.

2. Counting Procedure

The disintegration rate of each source was determined using the 4π β-(proportional)-γ coincidence technique with the gates in the gamma channel set on the 840-keV photopeak. Each source was measured to an accuracy of 0.05% (based on the total counts and the beta and gamma efficiencies). The measurements were started 5 h before the reference time and continued until 4 h after the reference time.

The average background corrections were 0.01, 0.06, and 0.001% for the beta, gamma, and coincidence channels, respectively, and the average deadtime and resolving-time correction was 0.5%.

The mean beta efficiency was 99.3%, and the mean gamma efficiency 13%. The decay-scheme correction was determined by measuring the apparent disintegration rate N_0' as a function of the beta efficiency. The beta efficiency was reduced by sandwiching the source between aluminum foils. The slope of the line N_0' versus $(1 - \epsilon_\beta)/\epsilon_\beta$ was $(4.5 \pm 2.5) \times 10^{-2}$.

3. Results

The value of the disintegration rate for each source was calculated using the resolving time and deadtime corrections given by Campion.[14] Since the average beta efficiency was 99.3%, no decay-scheme correction was made. A buoyancy correction of 0.1% was made to the source weights.

Measurements made on the same sources two days after the reference time showed that the effect of any contaminant on the beta and gamma count rates was less than 0.01% at the reference time.

The results are summarized in Table IX.

TABLE IX. NPL Data for [56]Mn Intercomparison

Reference Time: 2000 BST on September 11, 1969
Half-life: 2578 h

Ampoule No.:	No. at Reference Time in dps/mg			
	2	10	11	13
Source No.				
1	2523.51	2518.63	2520.37	2522.73
2	2524.29	2525.99	2523.66	2522.66
3	2522.11	2524.10	2529.10	2522.73
4	2528.51	2525.73	2521.14	2527.80
5	2522.81	2516.89	2526.77	2518.41
6	2529.02	2521.88	2520.70	2520.74
7	2522.40	2520.59	2520.81	2525.66
8	2526.06	2520.96	2524.84	2524.77
9	2528.04	2525.29	2522.29	2524.51
10	2527.84	2521.07		
Mean	2524.44	2522.11	2522.22	2523.33
% s.e.o.m.[a]	0.04	0.04	0.03	0.04
Overall mean	2523.03			
% s.e.o.m.	0.02			

Systematic Errors	%
Decay scheme	±0.03
Correction for accidental coincidences and deadtime	±0.05
Weighing	±0.05
Stability of measuring equipment	±0.05
Total systematic error	±0.18
Mean calibration figure for ionization chamber from these measurements	2.1574 pA/μCi ± 0.03%
Mean calibration figure for 30 previous standardizations	2.1577 pA/μCi ± 0.02%

(The quoted errors are the standard errors of the mean.)

[a] % s.e.o.m. = % standard error of the mean = $\sqrt{\dfrac{\sum(x_i - \bar{x})^2}{n(n-1)}} \times \dfrac{100}{\bar{x}}$.

ACKNOWLEDGMENTS

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